# Implications of particle composition and shape to dust radiative effect: A case study from the Great Indian Desert

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[1] The assessment of direct radiative forcing (DRF) of aerosol is uncertain, particularly where the natural dust particles mix with the anthropogenic components. One of the sources of such uncertainty is the assumption of morphology (size and shape) and composition of pure dust particles. Recently Mishra and Tripathi [2008] have computationally assessed the effect of particle morphology on optical properties over the Great Indian Desert. As a continuation of the previous study, in this paper, we have further examined the effects on dust radiative properties. Non-spherical pure dust particles show large variations in the optical and radiative properties from spherical pure dust particles, however, particle composition is found to have greater influence than particle shape on the radiative properties. Among the various shapes, sharp-edged particles show larger difference than smooth-shaped particles. Although the overall atmospheric absorption monotonically increases with increase in hematite content, maximum effect of particle non-sphericity at 4% hematite content implies that non-sphericity should be considered to minimize the uncertainty of regional estimates of aerosol DRF, as most of the global dusts contain that much hematite. However the difference in radiative properties for two different background dust cases due to particle morphology is low. Our results show that ignoring non-sphericity will lead to under-estimation of the regional warming and dust-absorption efficiency. **Citation:** Mishra, S. K., S. Dey, and S. N. Tripathi (2008), Implications of particle composition and shape to dust radiative effect: A case study from the Great Indian Desert, *Geophys. Res. Lett.*, *35*, L23814, doi:10.1029/2008GL036058.

## 1. Introduction

[2] Mineral dust has been identified as a key aerosol component in the Indo-Gangetic Basin, northern India, where it accounts for ~36% to the optical depth and ~40% to the surface aerosol direct radiative forcing (DRF) annually [*Dey and Tripathi*, 2008]. Mixing of pure dust particles with the anthropogenic pollution during the long-range transportation [*Chinnam et al.*, 2006; *Satheesh et al.*, 2006; *Dey et al.*, 2008] have been found to affect the optical and radiative properties in the region [*Dey et al.*, 2004; *Prasad et al.*, 2007]. Back trajectory analysis, satellite data and chemical evidence mark the Great Indian Desert (Thar Desert) as one of the major dust source regions, which increases the efforts to quantify the dust optical properties over there [*Moorthy et al.*, 2007]. The Great Indian Desert, an extensive arid region covering ~0.32 million km$^2$ of India and Pakistan, receives very little rainfall (<300 mm) annually and shows very high range of variation in surface temperature (~50$^0$ C in summer to ~−3$^0$ C in winter). Wind speed reaches 25–30 km hr$^{-1}$ in the summer raising tremendous amount of dusts in the atmosphere, and results in average soil loss of 30–60 kg m$^{-2}$ day$^{-1}$ in April–June [*Sikka*, 1997].

[3] As a first step to understand the dust radiative effects, accurate estimation of the optical properties of pure dust particles is essential. Desert dust particles are mainly composed of clay minerals, silicates (scattering components) and iron oxides (hematite is the main absorbing component) [*Koven and Fung*, 2006]. The dust particle size and shapes depend on many environmental factors, wind friction velocity and shear stress, soil roughness and moisture, atmospheric stability, presence of non-erodible elements, and cohesion of soil grains [*Mahowald et al.*, 2005]. *Parungo et al.* [1997] have observed that the relative abundance of particle shape varies in Asian dust, e.g. from 5% spherical, 70% sharp-edge and 25% smooth-shaped particles (Case 1) to 20% spherical, 50% sharp-edge and 30% smooth-shaped particles (Case 2). Their observations imply that non-sphericity is very important to account for the simulation of dust optical properties. Although, most of the studies have considered dust particles as spherical, recent advancement in simulation techniques [*Mishchenko et al.*, 2000] have allowed to consider the non-sphericity during the retrieval of dust optical properties from ground-based [*Dubovik et al.*, 2006] and satellite [*Kalashnikova et al.*, 2005] remote sensing.

[4] Efforts are being made to quantify the dust absorption using thermal infrared (TIR) satellite data [*Moorthy et al.*, 2007 and the earlier references therein] over the south Asian regions. This approach considers the satellite-measured TOA radiance at 10.5–12.5 $\mu$m wavelength region, from which the Infrared Difference Dust Index (IDDI) is derived and further used to estimate dust absorbing efficiency ($D_{AE}$) [*Deepshikha et al.*, 2005]. It is not precisely known how much the inferred spatio-temporal variation in the dust absorption is due to higher hematite content of pure dust or mixing with anthropogenic pollution or both. Examination of radiative effects of pure dust remains unexplored. *Mishra and Tripathi* [2008] have recently shown the effects of shape and composition on dust optical properties over the Great Indian Desert. The optical properties of pure dust have been simulated based on realistic particle shape and composition. As a continuation of the previous study, here we report the effect of the dust composition (in terms of

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absorption) and shape on the dust radiative effects. Our study also discusses the influence of particle morphology to TIR radiance.

## 2. Approach

[5] The detailed methodology of computing the optical properties of pure dust particles are described by *Mishra and Tripathi* [2008]. In brief, we have considered four different particle shapes, sphere, spheroid, cylinder and Chebyshev, based on the scanning electron microscopic analysis of the dust samples collected at Mt. Abu (a high-altitude station in the Aravalli range bordering the east of the Great Indian Desert). First, we have derived the index of refraction of dust particles. Although hematite is detected in the dust of the Thar Desert, its volumetric percentage is unknown. Hence, we have simulated our results by varying the hematite percentage from 0 to 10% based on the analysis of *Koven and Fung* [2006]. The refractive index of non-hematite part has been derived by volume-weighted average of individual refractive indices, the relative volume fractions have been taken from the study of *Peterson* [1968]. The effective wavelength-dependent complex refractive index for the dust has been calculated using the Bruggeman's effective medium mixing rule [*Bruggeman*, 1935].

[6] Optical properties (e.g. scattering and extinction cross-sections, asymmetry parameter ($g$), single scattering albedo (SSA) and scattering matrix elements) of the dust have been derived using T-matrix code [*Mishchenko and Travis*, 1998]. The optical properties of spherical particles have been derived using the Mie code by *Dave* [1968]. During the simulation, we considered particles in range 0.1–1 $\mu$m size with log-normal size distribution ($R_{mod}$ = 0.5 $\mu$m and $\sigma$ = 1.5) and aspect ratio of 1.5 [*Mishra and Tripathi*, 2008]. We have also considered two cases of background dust and computed the optical properties by taking weighted average of various individual shapes to show how they vary with shape. Optical properties of dust are then incorporated in the Santa Barbara Discrete Ordinate Radiative Transfer (SBDART) model [*Ricchiazzi et al.*, 1998] to compute the dust DRF at the top-of-atmosphere (TOA), surface and atmosphere for shortwave (0.25–4 $\mu$m) and TIR (10.5–12.5 $\mu$m) regions. The surface albedo has been considered as that of 'desert' model in SBDART. All the DRF estimations presented here are for clear-sky condition and diurnally averaged.

## 3. Results and Discussions

[7] The spectral variations of SSA and $g$ with the hematite content for cases 1 and 2 are displayed in Figure 1. Three important observations are evident from Figure 1. First is that the difference in spectral SSA and $g$ for any particular hematite content for case 1 and 2 is low (∼1–2% for SSA and ∼1–6% for $g$). Although the deviation of optical properties of individual shapes from spherical particles is higher, the combined effect (sometimes increasing and sometimes decreasing depending on hematite content and spectral variation) of various shapes is found to be low. Particularly for cylinders, the difference is very high, e.g. 5–7% for SSA, 2–7% for $g$ and ∼40% for extinction coefficient at 4–6% hematite content. These particles increase during the dust storms and thus regional estimates of aerosol radiative effects without considering non-sphericity may result in large uncertainty [*Kahnert et al.*, 2007]. Secondly, the spectral variability is high at higher hematite content, because the increasing concentration strongly governs the optical parameters (complex refractive indices, n and k) of the dust from ultraviolet to visible wavelength spectrum and the refractive index of hematite has more pronounced spectral dependence than the other components. Thirdly, the variation of SSA and $g$ with hematite content is much stronger at wavelength less than 0.6 $\mu$m, whereas it is practically non-existent at near-infrared (NIR) wavelength, because of the nearly non-absorbing nature of hematite at these wavelengths. Here SSA reduces by merely ∼2% with 10% increase in hematite content for both the cases. The reductions in SSA with increasing hematite content are higher at visible wavelengths (4–27%), and even more at ultraviolet (UV) wavelengths (∼48%). Sensitivity study by *Mishra and Tripathi* [2008] has shown that the optical properties of pure dust are more sensitive to hematite content than shape. Similar absorption may arise from pure dust with moderate (∼4–6%) hematite content and low-hematite dust mixed with anthropogenic black carbon. Note here that changes in SSA and $g$ also depend on particle size distribution [*Mishra and Tripathi*, 2008]. The highest difference in visible SSA due to particle shape was found to be ∼4%. Spectral variation of $g$ on the other hand shows a dip at the visible wavelength (minimum at 0.6–0.7 $\mu$m) for both the cases. The absolute values of $g$ increase with higher hematite content at UV and VIS (up to 0.6 $\mu$m) and show reverse trend at wavelengths higher than that. Higher absorption reduces the contribution of internal resonances inside the particles resulting in less side-scattering and thus increasing $g$ with increasing hematite content at shorter wavelengths. At longer wavelength, decrease in prevailing forward scattering and much lesser influence of hematite absorption result in reverse behavior of $g$. Here it should be mentioned that more realistically deviation of $g$ from that of spherical particles due to non-sphericity depends not only on refractive index, but also on the size parameter and surface roughness effects [*Kahnert and Nousiainen*, 2006; *Rother et al.*, 2006; *Mishchenko et al.*, 1997].

[8] The observed changes in the optical properties of pure dust due to shape and composition reflect on the change in dust DRF. As seen from the optical properties, the DRF values expectedly show very small difference (<2%) for these two cases and the influence of composition is also very similar (not shown). This means to account non-sphericity in the regional estimates of DRF, any proportion of non-spherical particles between these two cases may be used without much difference. Dust DRFs at TOA and surface vary between −14 to −6 W m$^{-2}$ and −15 to −28 W m$^{-2}$. Higher hematite content in dust particles causes lesser cooling at TOA and higher cooling at surface, effectively increasing the atmospheric absorption manifold. Atmospheric heating rate due to pure dust alone at 4% hematite content is around 0.4 K day$^{-1}$, similar to the values reported by *Satheesh et al.* [2007] that were estimated from the dust optical properties derived from $D_{AE}$ images. If we consider the individual shapes, the SW dust DRF shows much higher variability. The atmospheric absorption is

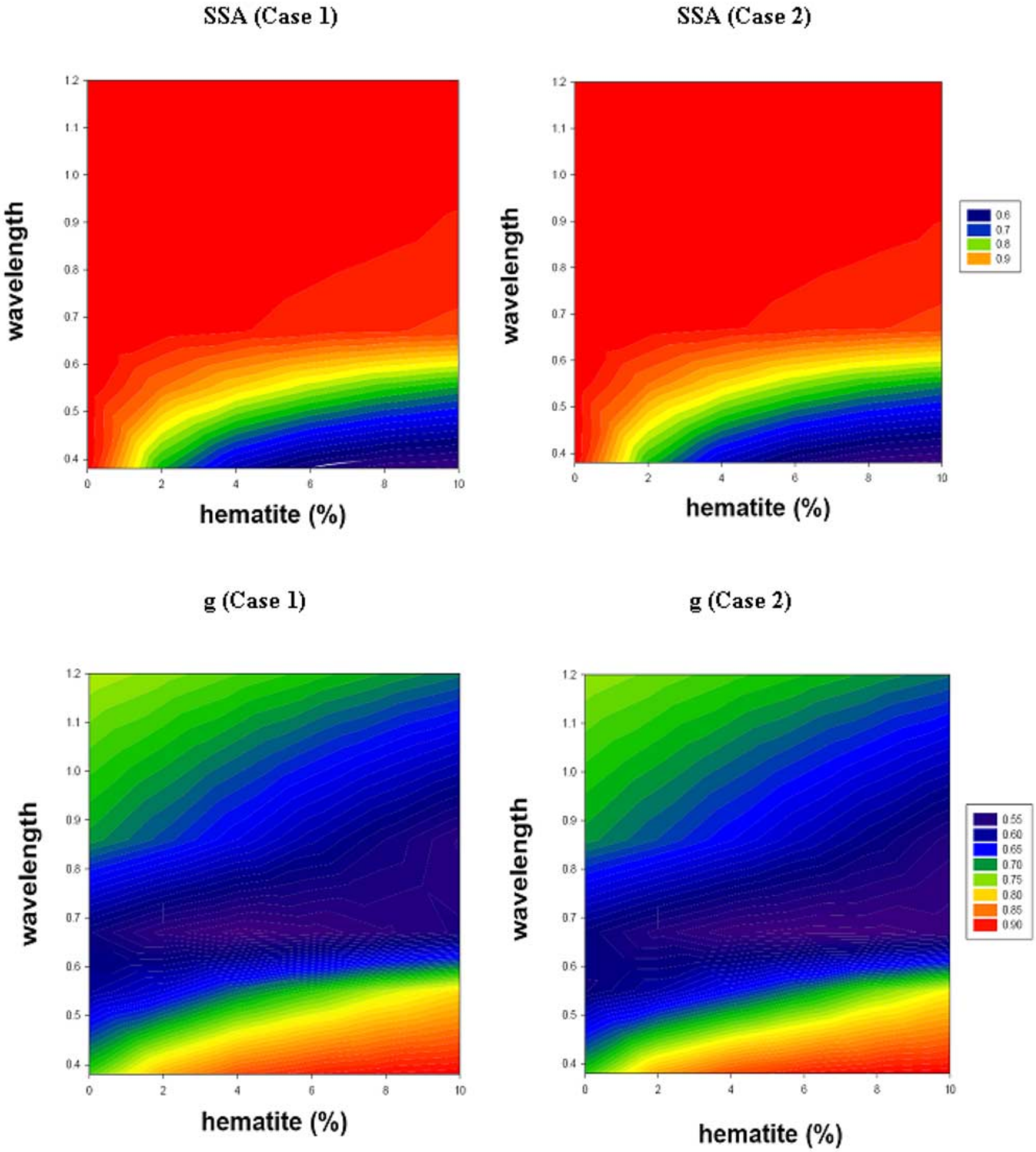


**Figure 1.** Spectral variation of (top) SSA and (bottom) g for (left) case 1 and (right) case 2 of 'background dust' with varying hematite percentage. The red color indicates SSA (top) higher than 0.95.

almost negligible ($\sim$2.5 W m$^{-2}$) without hematite irrespective of shapes, but drastically increases manifold at only 2% hematite content suggesting the strong influence of hematite on absorption characteristics. Percentage departure of DRF values for non-spherical particles from that for spherical particles is shown in Figure 2 to judge the effect of only particle morphology on DRF with other parameters (such as surface albedo, water vapor etc.) remaining constant. The cylindrical particles show highest deviation from the spherical particles followed by the Chebyshev and spheroid particles with a peak at 4% hematite content. Clearly the non-sphericity attributes to more cooling at both TOA and surface, and combined effect is $\sim$6% more warming than the spherical particles. The smooth-shaped particles show similar kind of deviation from the spherical particles due to similarity in their scattering phase functions.

[9] Consideration of more complicated, sharp edged and non-axisymmetric particles will give rise to significant

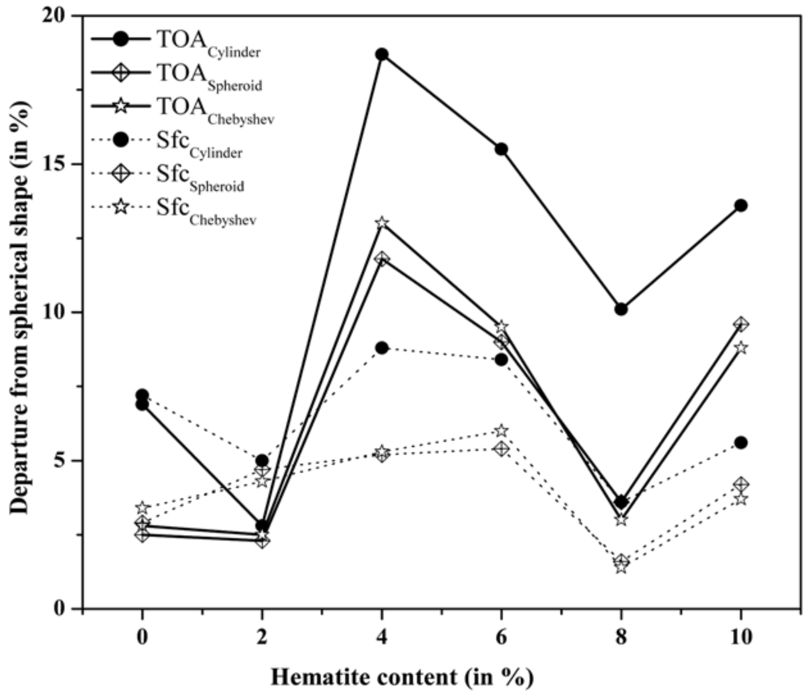


**Figure 2.** Percent departure of dust DRF at TOA and surface (Sfc) due to non-spherical shapes (in subscript) from dust DRF due to spherical shape with varying hematite content.

deviation from their volume equivalent spherical counterpart in forcing estimation. But given the limitation of the computational method to treat those particles, even the present calculations show large influence of shape on dust DRF. *McComiskey et al.* [2008] have shown that optical depth and SSA have strongest sensitivity on the DRF, and these optical properties show large deviation with shape and composition [*Mishra and Tripathi*, 2008]. The increase of sharp edged particles with hematite content >2% during the dust storms will give rise to more heating of atmospheric column. The significance of consideration of particle shape is more in the regions with high hematite content (>5%), i.e. east and middle-east Asia [*Koven and Fung*, 2006] and the regions where black carbon mixes with pure mineral dust (i.e. Indo-Gangetic basin in northern India and East Asia), because enhancement in the atmospheric warming will be under-estimated if particle morphology is not considered.

[10] The uncertainty in the recent estimates of aerosol DRF over the Indo-Gangetic basin [*Dey and Tripathi.*, 2008] will be minimized if the aerosol model is improved by incorporating the dust particle morphology, because the region suffered frequent dust storms from the Great Indian Desert and middle-east Asia. Particle non-sphericity is also important as *Mishchenko et al.* [1995] have shown that the consideration of tropospheric dust aerosols as spherical particles while retrieving aerosol optical thickness from measured satellite reflectance leads significant error in the retrieval. The error in the TOA radiative forcing estimation for feldspar aerosol lies between −6 and 31% when simple spherical model particles have been considered in the computations [*Kahnert et al.,* 2005] whereas the consideration of the moderate non-sphericity i.e. spheroid particle shape shrinks the error range to −1 to 6%.

[11] We have examined the influence of particle shape and composition on the variation of the infrared brightness temperature (IRBT). Due to limitation in the T-matrix code, the optical properties computed up to 1.2 $\mu$m wavelength are extrapolated to the LW region and then used to compute the LW fluxes. The TOA outgoing flux due to dust and non-dust cases at the 10.5–12.5 $\mu$m wavelength is converted to IRBT using inverse Planck function [*Satheesh et al.*, 2006]. The difference in IRBT for the dust and non-dust case, defined as IDDI [*Deepshikha et al.*, 2005], has been calculated for all the cases discussed above. The variation of IDDI due to particle shape and composition is less than 2%, but at the same time, increase in extinction coefficient will lead to an increase in aerosol optical depth. The difference could be larger for coarser particles [*Mishra and Tripathi*, 2008]. As $D_{AE}$ is a ratio of IDDI and AOD [*Deepshikha et al.*, 2005], under-estimation of AOD will lead to under-estimation of $D_{AE}$, given that IDDI does not change much for an aerosol system predominantly composed of mineral dust (e.g. contribution of dust is more than 60% to AOD during premonsoon season in the northern India [*Dey and Tripathi*, 2008]). This influence will obviously be less for the time when relative contribution of dust reduces (e.g. dust contributes ~18% to AOD in winter season), but even then it can not be neglected.

## 4. Summary and Conclusions

[12] In the present paper, the effects of particle nonsphericity and composition on the pure dust DRF over the Great Indian Desert have been examined. The radiative transfer computations account the optical properties of pure dust for four major particle shapes (spherical, spheroid, cylinder and Chebyshev) found in the region and varying

composition of hematite content within the range 0–10% reported by *Mishra and Tripathi* [2008]. It should be noted here that if more complex and non-spherical shapes can be considered, the results may show larger deviation from spherical particles and larger absorption, but ability to treat more complex shape is computationally limited. Based on the simulations, the major conclusions are as follows.

[13] 1. Particle composition has stronger effect than the shape on the integral radiometric characteristics of pure dust. Absorption increases (as indicated by decrease in SSA) at higher rate at UV than VIS and is negligible at NIR wavelength with increase in hematite content.

[14] 2. Dust DRF shows large variation due to shape and composition with sharp-edged cylindrical particles showing higher deviation than the smooth-shaped particles from spherical particles. Maximum deviation of DRF from that of spherical particles at around 4% hematite content implies that non-sphericity must be considered for accurate estimation of regional aerosol radiative effects, as most of the global dust source regions have hematite content around or more than that value Consideration of spherical particles in the regional estimates of aerosol DRF will lead to underestimation of the warming.

[15] 3. Ignoring the particle morphology will lead to under-estimation of optical depth, and subsequently underestimation of $D_{AE,}$ as IDDI is not sensitive to particle shape.

[16] While the present results show the importance of consideration of particle morphology to assess regional aerosol DRF more accurately, few questions still unresolved should be considered in future. It is important to determine the hematite content of the Indian dusts to narrow down the range of variations of the optical properties. The variation of the optical and radiative properties of non-spherical shapes with size distribution should be examined quantitatively. Furthermore, radiative effects of dust-black carbon mixture in this region so far have been studied using spherical dust particles [*Dey et al.*, 2008; *Satheesh et al.*, 2006]. Treatment of non-sphericity in dust-black carbon mixture is another important aspect for future investigations.

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